PB-217 642

TRITIUM CONCENTRATION IN PRECIPITATION, 1967-1968

A. A. Moghissi, et al

Environmental Health Service
Rockville, Maryland

1969

DISTRIBUTED BY:

NTIS

National Technical Information Service
U. S. DEPARTMENT OF COMMERCE
5285 Port Royal Road, Springfield Va. 22151

PB 217 642

Reprinted by
U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service
Environmental Health Service
From Radiological Health Data and Reports, Vol. 11, Number 3, March 1970

Tritium Concentration in Precipitation, 1967-1968

A. A. Moghissi and *C. R. Porter*[1]

Since the discovery of natural tritium (*1–2*), rain has been extensively monitored to study the production and behavior of this radionuclide in the hydrological cycle (*3*). Although the methodology of early tritium studies (*4*) proved to be somewhat unreliable (*5*), these findings are the basis of our present knowledge regarding the natural concentration and seasonal variation of tritium.

It has been well established that under normal conditions—i.e., in the absence of the introduction of tritium by nuclear bomb testing—tritium concentration in rain will increase during spring and late summer in the Northern Hemisphere (*3*). In the Southern Hemisphere the conditions are similar, although less pronounced (*3*).

Because of the mode of production of natural tritium (*1–3*), it may be expected that the tritium concentration will increase in higher altitudes. Also, numerous studies have shown that tritium concentrations reach a maximum at 45–60° N latitude in the Northern Hemisphere (*3*). There is presently no adequate explanation for this phenomenon.

Rain analysis represents a sensitive technique for monitoring of tritium in the environment. Any significant addition of tritium into the natural cycle can easily and rapidly be detected by rain analysis, primarily due to considerable improvements in the low-level counting techniques for tritium (*6*).

[1] Dr. Moghissi was at Southeastern Radiological Health Laboratory, Montgomery, Ala. and now is with Southwestern Radiological Health Laboratory, Las Vegas, Nev. 89114. C. R. Porter is director, Southeastern Radiological Health Laboratory, Montgomery, Ala. 36101.

Beginning in 1967, the Bureau of Radiological Health established a tritium in rain network as part of the existing Radiation Alert Network. Ten stations were selected covering the United States, including Alaska and the Hawaiian Islands. The locations of these stations are shown in figure 1.

Procedure

Rain samples from the collection sites are shipped to Rockville, Md., where they are composited on a monthly basis. Part of each monthly composite is transferred to disposable containers and shipped to the Southeastern Radiological Health Laboratory where they are distilled and counted by a liquid scintillation technique. The details of the analytical procedure have been described elsewhere (*7*). It consists of the addition of 20 ml of a scintillation liquid to 5 ml of water in a plastic vial. The scintillation liquid consists of spectrographic grade dioxane containing 7 g diphenyloxazole (PPO), 1.5 g *p*-bis-(o-methylstryl)-benzene(bis-MSB), and 120 g naphthalene per liter. All operations involving the scintillation solution are carried out under red light. Plastic vials, manufactured by Nuclear Chicago Corporation, and a Beckman liquid scintillation counter are used. Under these conditions, a counting efficiency of 22.5 percent was obtained with a background of 8.0 cpm resulting in a sensitivity of 0.40 nCi/liter-cpm.

Duplicate samples were prepared and counted twice for 50 minutes. A background sample was counted before the first sample and a second background after the duplicate sample. If any one of

Reproduced by
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. Department of Commerce
Springfield VA 22151

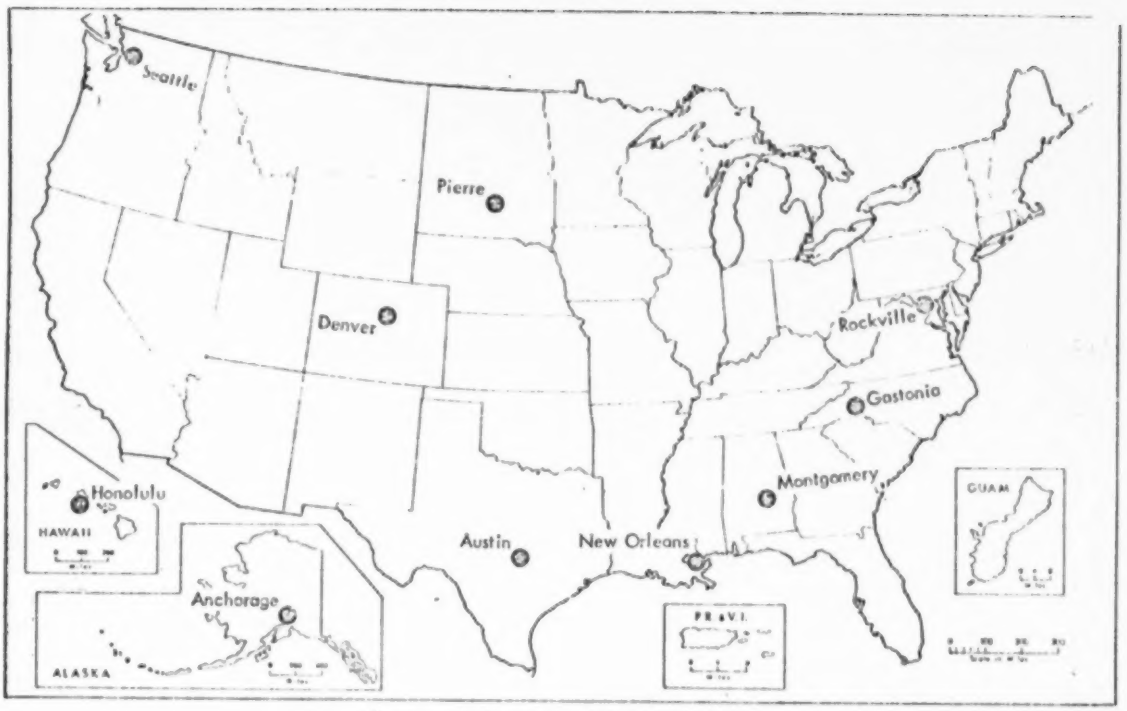

Figure 1. Tritium precipitation stations

Results

Tables 1 and 2 show the analytical results obtained from the samples collected during 1967 and

the 50-minute sample counts deviated by more than 2-sigma from the average, the duplicates were recounted. This occurred, however, with a frequency comparable to the statistical expectation. The error of the analysis at the 2-sigma confidence level as a function of activity is shown in figure 2. The minimum limit of detection was 0.2 nCi/liter at the 2-sigma confidence level.

The results of this investigation are indicative of radiological health implications of tritium in the atmosphere. It can be clearly seen that tritium in the atmosphere is continuously decreasing as compared to 1965 samples (*3*). Tritium concentrations in rain for previous years are not available from the same geographical locations of this investigation. It is, however, interesting to consider that levels as high as 6 nCi/liter were reported for the continental United States during 1965.

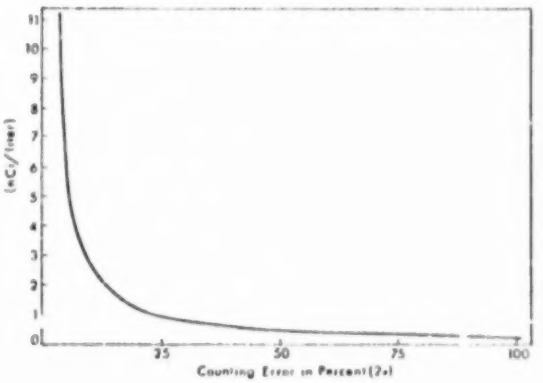

Figure 2. Correlation between tritium concentration of water and counting error in percent at the 2-sigma confidence level

Table 1. Tritium analysis of rain composites from RAN stations, 1967

Location	Concentration (nCi/liter)											
	Jan	Feb	March	April	May	June	July	Aug	Sept	Oct	Nov	Dec
N.C. Gastonia	0.2	0.5	0.5	0.6	0.3	0.2	0.3	<0.2	0.8	<0.2	<0.2	0.3
Md. Rockville	(*)	.6	.7	1.0	.7	.5	(*)	(*)	(*)	<.2	<.2	<.2
Ala. Montgomery	(*)	.5	<.2	(*)	(*)	<.2	<.2	(*)	.9	1.2	<.2	<.2
S.Dak. Pierre	(*)	(*)	(*)	2.0	2.1	1.3	(*)	(*)	<.2	(*)	(*)	(*)
La. New Orleans	<.2	<.2	.4	<.2	<.2	<.2	.3	<.2	.4	<.2	<.2	<.2
Tex. Austin	<.2	<.2	.5	<.2	<.2	(*)	<.2	<.2	<.2	<.2	<.2	<.2
Colo. Denver	.5	1.0	1.1	2.6	1.7	1.4	1.4	1.1	1.2	.3	.5	.4
Hawaii Honolulu	<.2	.2	.2	.4	<.2	(*)	<.2	<.2	1.4	.3	<.2	<.2
Wash. Seattle	<.2	.4	.7	.6	.7	.5	(*)	(*)	1.6	<.2	<.2	<.2

* Sample not received.

Table 2. Tritium analysis of rain composites from RAN stations, 1968

Location	Concentration (nCi/liter)											
	Jan	Feb	March	April	May	June	July	Aug	Sept	Oct	Nov	Dec
N.C. Gastonia	0.4	<0.2	<0.2	0.5	0.2	0.5	<0.2	(*)	<0.2	<0.2	<0.2	<0.2
Md. Rockville	<.2	(*)	<.2	(*)	(*)	(*)	(*)	<0.2	(*)	<.2	(*)	(*)
Ala. Montgomery	<.2	<.2	.3	.5	.2	.5	<.2	<.2	<.2	<.2	<.2	<.2
Alaska Anchorage	(*)	(*)	(*)	(*)	.5	.5	.7	.7	<.2	<.2	(*)	<.2
La. New Orleans	.2	<.2	<.2	<.2	<.2	<.2	<.2	<.2	<.2	(*)	(*)	(*)
Tex. Austin	<.2	.9	<.2	<.2	<.2	<.2	<.2	<.2	<.2	<.2	<.2	(*)
Colo. Denver	.4	1.1	.5	1.7	1.2	.6	.5	.4	.6	<.2	.5	(*)
Hawaii Honolulu	<.2	<.2	<.2	<.2	(*)	<.2	(*)	(*)	.5	(*)	<.2	(*)
Wash. Seattle	.2	<.2	<.2	<.2	.3	<.2	<.2	.3	<.2	<.2	<.2	<.2

* Sample not received.

1968. The station at Montgomery, Ala., is not a sample collection site; but due to the availability of this sample, it was run for comparison.

The spring and late summer peaks are clearly detectable in the majority of cases. The station with the highest tritium level is Denver, Colo. Figure 3 shows the tritium concentration of rain samples during the entire sampling period from this station. The reason for the relatively high levels of tritium in rain from Denver is attributed primarily to the high altitude of this location.

The effect of latitude is shown in samples collected from Pierre, S. Dak., from which, unfortunately, only three samples were received. This was the only station within the 45-60° N latitude that

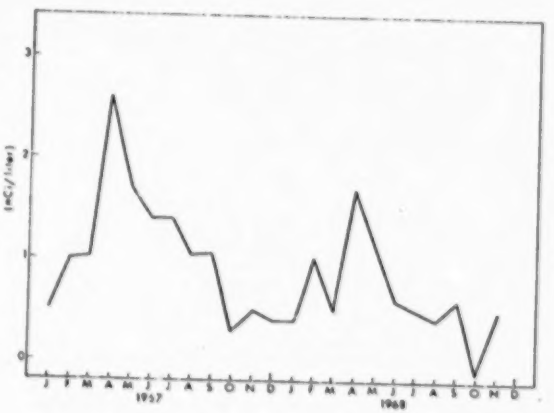

Figure 3. Tritium activity of precipitation in Denver, Colo., during 1967–1968

would not be affected by dilution from ocean evaporation.

The analysis of samples received during the last 2 months of the sampling period reported was carried out using a suspension technique (8) with a minimum limit of detection of 0.1 nCi/liter. As this was in an experimental state and for the sake of uniformity, these values are reported to the same sensitivity as the previously used method.

The analytical technique used, although a sensitive method, is inadequate for any geophysical prediction. An adequate technique for the observation of tritium concentration in rain for this purpose would require a minimum limit of detection of 10–20 pCi/liter of water.

Acknowledgment

The technical assistance of Mae W. Williams and Rachael H. Gorrie in the preparation of this paper is greatly appreciated.

Mention of commercial products used in connection with work reported in this article does not constitute an endorsement by the Public Health Service.

REFERENCES

(1) FALTING, V. and P. HARDECK. Tritium content of the atmosphere. Z Naturforsch 5a:438 (1950).
(2) GROSSE, A. V., W. H. JOHNSTON, R. L. WOLFGANG, and W. F. LIBBY. Tritium in nature. Science 113:1 (1951).
(3) INTERNATIONAL ATOMIC ENERGY AGENCY. Tritium and Other Environmental Isotopes in the Hydrological Cycle, Technical Report No. 73. International Atomic Energy Agency, Vienna (1967).
(4) KAUFMAN, S. and W. F. LIBBY. Natural distribution of tritium. Phys Rev 93:1337 (1954).
(5) SCHUMACHER, E. Methodology for tritium in natural waters. Helv Chim Acta 43:1019 (1960).
(6) MOGHISSI, A. A. Radionuclides in the Environment. Analysis and Public Health Aspects of Environmental Tritium. Advances in Chemistry Series, American Chemical Society, Washington, D.C. (in press).
(7) MOGHISSI A. A H. L. Kelley, J. E. REGNIER and M. W. CARTER. Low-level counting by liquid scintillation. 1. Tritium measurement in homogeneous systems. Inter J Appl Rad Isotopes 20:145 (1969).
(8) LIEBERMAN, R. and A. A. MOGHISSI. Low-level counting by liquid scintillation. 2. Application of emulsions in tritium measurements. Inter J Appl Rad Isotopes (in press).